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(54) Sealing Compound for Producing Multilayer Insulating Glass

(57) A sealing compound for producing multilayer insulating glass containing silane-modified butyl rubber and used as a spacer between the panes of glass of the multilayer insulating glass is described.

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Description

The present invention relates to a new sealing compound which is suitable for producing multilayer insulating glass.

Insulating glass consists of at least two panes of glass separated by a spacer. The spacer is placed in the edge area of the panes of glass. This arrangement is usually sealed by a combination of two different sealing compounds. By thermal and acoustic separation of the two panes of glass, a component that provides both thermal and noise insulation is obtained; the properties of this component can be further improved by filling it with various gases (e.g., noble gases such as argon, krypton or xenon or heavy gases such as sulfur hexafluoride) and especially by using coatings on the glass.

The type of spacer used with conventional commercial insulating glass arrangements is a hollow profile made of metal (preferably aluminum, galvanized steel or stainless steel) or made of plastic (e.g., polycarbonate) which can accommodate in the hollow space a granular desiccant for adsorption of moisture and possibly even solvent residues, in which case the hollow space is connected to the enclosed volume between the panes of glass through small openings and can keep the glass dry. The spacer is positioned on the glass in such a way that it forms a joint with the edge areas of the panes of glass.

The sealing compounds generally used for multilayer insulating glass are classified in two categories: a primary sealing compound ("butyl") is used to seal the unit to prevent atmospheric humidity from penetrating into it and to prevent the escape of the filling gases; such a primary sealant is based on isobutylene polymers which have a low water vapor permeability. It is provided between the glass and the surface of the spacer facing toward the glass. A second sealing compound which is used for elastic bonding usually contains silanes for improved adhesion to glass and must withstand the stresses caused by all climate conditions; this second sealant fills the joint formed between the panes of glass and the spacer. The second sealant usually consists of two-component sealing compounds that cure at room temperature such as polysulfide, polyurethane or silicone. Another conventional variant is to fill the joint with a hot-melt adhesive based on butyl rubber ("butyl hot melt"). In this case, the use of a primary sealant may be omitted because the butyl hot melt may be used for general sealing purposes because it already has a good imperviousness with respect to water vapor.

A major disadvantage of the production of insulating glass by the method described previously is the use of a plurality of materials which must be processed in a number of operations in a cost-intensive process. A first step toward better automation of the insulating glass production was made with the development of the so-called "Biver system" or the new "TPS system" as described in the European Patent Application EP A-0714964, for example. In this system, a strand of a thermoplastic material is extruded onto a first pane of glass by means of a suitable machine; the second pane of glass is placed over the first and then pressed to a predetermined spacing. As in the previous case, the joint which is also formed in this process between the strand

based on isobutylene polymer and the glass is filled with a traditional two-component sealing compound that hardens at room temperature and serves to form an elastic bond. The internal strand of the thermoplastic material not only assumes the function of a "spacer" but also serves as a water vapor and gas barrier and serves as a desiccant.

This insulating glass manufacturing system has the great disadvantage that elastic sealing of the multilayer insulating glass units requires the use of a two-component sealing compound that cures at room temperature, so this means that there are rinsing losses and downtimes for the sealing equipment and there may even be a "waxy blockage" of the sealing apparatus due to slow curing of the material residues remaining in the mixer. Another disadvantage is that a certain waiting time must be observed; within this waiting period the two-component sealing compound must crosslink to a certain minimum hardness before the multilayer insulating glass units can be stacked, packaged and shipped.

Japanese Patent Application JP A-09086975 describes a method for producing a flexible, self-stick rubber-like spacer for double layer insulating glass with which a spacer is produced from a mixture of a partially crosslinked butyl rubber with an organosilicon compound, a desiccant and additional ingredients (tackifier, reinforcing and non-reinforcing fillers). One disadvantage of this method is that it yields merely a physical mixture of the components in which the incorporated organosilicon compound functions only as a primer when applied to glass and no chemical bonding to the polymer (partially crosslinked butyl rubber) is possible.

Unexamined German Patent DE-A-196 24 236 discloses a hot-melt adhesive composition for producing insulating glass without the use of additional spacers; this composition is a mixture of silane-functional polyisobutylenes, silane functional hydrogenated polybutadienes and/or silane-functional poly-α-olefins with non-reactive polymers such as rubbers, poly-α-olefins or polybutenes. One disadvantage is that the hydrogenated polybutadienes and poly-α-olefins that are used have only limited suitability as the main components of the mixture due to their high water vapor permeability; secondly, synthesis of the silane-functional polyisobutylenes can take place only in a tedious multi-step process starting with olefinically terminated or hydroxyl-terminated telecheles (oligomers with two functional end groups) by reaction with suitable organofunctional silanes. The possibility of grafting (reaction between a polymer and another molecule that is usually small, forming a polymer with a side group on the main chain) is also mentioned in this publication; in this case the reaction of organofunctional silanes with nonfunctional polyisobutylenes cannot be implemented because the saturated hydrocarbon chains of isobutylene units disintegrate into shorter chain fragments in the reaction (see, for example, K. Kircher, *Chemische Reaktionen bei der Kunststoffverarbeitung* [Chemical Reactions in Plastics Processing], Carl Hanser Verlag, Munich, Vienna 1982, p. 218).

In the Patent Application EPA-A-0 312 967, a sealing compound for double layer insulating glass with a metal spacer is described; this sealing compound contains a polymer with a relatively low molecular weight (500 to 30,000 g/mol) which is composed of a saturated hydrocarbon with at least one crosslinkable group containing silicon. Such polymers with a molecular weight of 1000 to 15,000 g/mol are preferred. It is a disadvantage here that because of the low molecular weight of the crosslinkable polymer, it is indeed possible to obtain low-

viscosity pumpable sealing compounds that are suitable for sealing traditional multilayer insulating glass having a metallic spacer, but they do not appear to be suitable for production of insulating glass units without additional spacers. In addition, synthesis of the silane-modified polyisobutylenes and hydrogenated polybutadienes described here requires a great expenditure for equipment using solvents and working under a protective gas atmosphere.

Therefore, the object of this invention is to overcome the disadvantages of the state of the art and to develop a novel sealing compound for sealing multilayer insulating glass with which insulating glass can be produced inexpensively, while eliminating several operating steps, and in addition it may be used as a spacer in particular.

This object is achieved by a sealing compound containing a silane-modified butyl rubber. Features of the inventive sealing compound are characterized in Claims 1 through 12. Claims 13 through 18 characterize the use of the inventive sealing compound.

It has surprisingly been found that butyl rubber can be modified easily with crosslinkable silanes containing mercapto groups and that this modified butyl rubber is suitable for inexpensive production of a sealing compound with which multilayer insulating glass can be produced without requiring an additional spacer.

The inventive sealing compound consists of silane-modified butyl rubber and additional ingredients. The respective amounts (in wt%) of the various components are listed below:

(a) silane-modified butyl rubber	5-70%
(b) thermoplastic polymer	5-40%
(c) rubbers	0-40%
(d) tackifying additives	5-50%
(e) crosslinking catalysts	0-2%
(f) adhesion promoter substances	0-5%
(g) stabilizers	0.1-4%
(h) reinforcing and non-reinforcing fillers	5-60%
(i) desiccants	1-40%

The inventive sealing compound preferably has the following composition:

(a) silane-modified butyl rubber	10-60%
(b) thermoplastic polymers	7-35%
(c) rubbers	2-35%
(d) tackifying additives	6-40%
(e) crosslinking catalysts	0-1%
(f) adhesion promoter substances	0-3%
(g) stabilizers	0.2-3%
(h) reinforcing and non-reinforcing fillers	7-50%
(i) desiccants	5-35%

Butyl rubber is a commercially available polymer consisting of isobutylene and isoprene monomer units, where the isoprene units are present only in a small amount, usually up to 3 mol% in the polymer (examples include various grades of butyl from the companies Bayer AG, Exxon Chemicals and Kautschuk Gesellschaft [Rubber Company]).

Partially crosslinked grades of butyl rubber with a degree of crosslinking of up to 80% are available commercially and are accessible to functionalization; they are obtainable, e.g., by polymerizing a bifunctional monomer (e.g., divinylbenzene) or by subsequent crosslinking of butyl rubber (examples include LC butyl from Exxon Chemical, Kalar from Hardman and Polysar Butyl XL from Bayer).

The silane functionalization of the butyl rubber is achieved by reacting the solid butyl rubber (molecular weight >30,000 g/mol) at an elevated temperature in a suitable mixing unit, e.g., a compounder or a single-screw extruder or a twin-screw extruder with a silane that contains mercapto groups and is crosslinkable via hydroxyl groups or hydrolyzable groups and with a free radical forming agent. Elevated temperature here means a temperature close to or above the decomposition temperature of the free radical forming agent which may decompose into free radicals (splitting off a volatile component such as nitrogen in some cases). These free radicals cause abstraction of a hydrogen atom from the mercapto group of the silane that contains mercapto groups, forming a sulfur radical. This sulfur radical can then react with the original isoprene unit within the main chain of the butyl rubber by substitution on the methylene group that is vicinal to the double bond or by addition at the double bond, thereby permitting covalent bonding of the silane that contains mercapto groups to the polymer.

It is advantageous here that no additional instrument expense is necessary for this reaction. The reaction can be carried out in the same equipment in which the other ingredients are combined to form the sealing compound according to this invention.

The silane that contains mercapto groups can be described by the following general formula:

[see source for formula]

wherein
A denotes $(CH_2)_m$ or R^3-NH-CO-NH-R^4 or R^3-O-R^4 or R^3-S-R^4 and
m stands for a number between 1 and 10, preferably 1 to 6, especially preferably 3, and
R^1, R^2 stands for a substituted or unsubstituted alkyl group with 1 to 20 carbon atoms or a substituted or unsubstituted aryl group with 6 to 20 carbon atoms or a substituted or unsubstituted aralkyl group with 7 to 20 carbon atoms and
R^1 and R^2 may be the same or different and
X stands for hydroxyl group or a hydrolyzable group and when there are several X groups in a molecule, they may be the same or different and
a stands for 0, 1 or 2 and
b stands for 0, 1, 2 or 3 and
the sum of a and b is no greater than 4 and
n is an integer between 0 and 19 and

R^3, R^4 stand for $(CH_2)_m$ or $(CH_2)_m$-NH-$(CH_2)_m$ and
R^3 and R^4 may be the same or different.

Examples for the hydrolyzable group X include a hydrogen atom, an alkoxy group, an acyloxy group, a ketoximate group, an amino group, and amido group, an aminoxy group, a mercapto group or an alkeneoxy group. Of the groups listed here, the alkoxy groups are preferred because they undergo hydrolysis under mild conditions and are easy to handle.

Preferred silanes containing mercapto groups include:

[see source for formula]

3-mercapto-propyl-trimethoxysilane (brand names: Dynasylan MTMO (Sivento), A-189 (OSi), Sila-Aoe S 810 (Chisso) and Silane GF 70 (Wacker)):

[see source for formula]

3-mercapto-propyl-triethoxysilane (brand names: Dynasylan 3201 (Sivento), A-1891 (OSi));

[see source for formula]

3-mercapto-propyl-dimethoxysilane (brand names: Dynasylan 3403 (Sivento), Silane SLM 50970 (Wacker)).

The free radical forming agent necessary for the graft reaction can be selected from the group of azo compounds such as azonitriles (e.g., 2,2'-azobisisobutyronitrile), azoamides (e.g., 1,1'-azo-bis-formamide), azoalkanes, azoalkenes, azoalkynes or azoaryls (e.g., 2,2'-azobispropane, 2,2'-azobis-2-methylbutane, methyl-azo-3-propene, azo-bis-(1,1-dimethyl)-2-propyne, 1,1-azo-bis-cumene, phenyl-azo-triphenylmethane) or azocarboxylates (e.g., dimethyl-2,2'-azo-isobutyrate), from the group of peroxide compounds such as alkyl peroxides (e.g., di-*tert*-butyl peroxide or dicumyl peroxide), acyl peroxides (e.g., acetyl peroxide, propionyl peroxide or dibenzoyl peroxide), hydroperoxides (e.g., *tert*-butyl hydroperoxide), per esters (e.g., *tert*-butyl peracetate or *tert*-butyl perisobutyrate) or peroxycarbonates (e.g., diisopropyl peroxydicarbonate) as well as from the group of redox initiators (e.g., the combinations hydroperoxide/Fe (II), hydroperoxide/Cu (II), hydroperoxide/Cu (I) (where a hydroperoxide can be described by the formula ROOH, where R = alkyl group with 1 to 20 carbon atoms or R = aryl group with 6 to 20 carbon atoms or R = aralkyl group with 7 to 20 carbon atoms) or the combination hydrogen peroxide/Fe(II)) or from the group of other initiators that disintegrate into radicals. The graft reactions can be performed to advantage with 2,2'- azobisisobutyronitrile, di-*tert*-butyl peroxide, dicumyl peroxide and dibenzoyl peroxide.

The composition temperature of the free radical forming agent is in the range of less than 0°C to more than room temperature in the case of the redox initiators and in the case of the other initiators mentioned as examples, the decomposition temperature is above room temperature. For

example, 2,2'-azobisisobutyronitrile decomposes at more than 50°C and dicumyl peroxide at more than 82°C. The reaction of butyl rubber with a silane that contains mercapto groups and a free radical forming agent is carried out in a suitable apparatus, preferably in a compounder, a single-screw extruder or a twin-screw extruder at temperatures up to approx. 220°C. The reaction temperature is preferably between 80°C and 220°C.

Instead of butyl rubber, a butyl rubber that is already partially crosslinked may also be used as the starting material for production of the silane-modified butyl rubber.

In addition to the silane-modified butyl rubber, the sealing compound also consists of other ingredients which improve the properties of the sealing compound. These ingredients include thermoplastic polymers, natural and synthetic rubber, tackifying additives, crosslinking catalysts which catalyze the hydrolysis and crosslinking of the silane groups, adhesion promoter substances such as silanes or metallates, stabilizers such as antioxidants, reinforcing and non-reinforcing fillers, desiccants.

Examples of thermoplastic polymers include polyolefins as homopolymers or copolymers composed of the monomers ethylene, propylene, *n*-butene and their higher homologs and isomers, functional vinyl compounds such as vinyl acetate, vinyl chloride, styrene, α-methylstyrene and unsaturated acid derivatives such as acrylic acid, methacrylic acid, acrylates, methacrylates and acrylonitrile. Other examples include polyamides [nylons], polyimides, polyacetals, polycarbonates, polyesters and polyurethanes as well as blends of all the polymers mentioned above.

Natural and synthetic rubbers may be selected from the group of homopolymers of dienes and the group of copolymers and terpolymers of dienes with olefins. Examples include polybutadiene, polyisoprene, polychloroprene, styrene-butadiene rubber, block copolymers with blocks of styrene and butadiene or isoprene, butyl rubber, high-molecular polyisobutylene, ethylene-propylene rubber and ethylene-propylene-diene rubber, e.g., with dicyclopentadiene or ethylidene norbornenes as the diene component. The rubber may also be used in hydrogenated form or in blends. However, the sealing masses according to this invention may also be produced without rubber.

Tackifying additives may be selected from the group of natural and synthetic resins including those that are modified subsequently and include, for example, hydrocarbon resins, colophony and derivatives thereof, polyterpenes and derivatives thereof, coumarone-indene resins, phenolic resins and acrylate resins and from the group of polybutenes, polyisobutylenes and degraded liquid rubbers (e.g., butyl rubber or EPDM) which may also be hydrogenated. Mixtures of the tackifying additives listed above may also be used.

Crosslinking catalysts may include all reagents that catalyze the hydrolysis of the silanes containing mercapto groups and the subsequent crosslinking reaction by condensation of the silanol groups. Especially preferred examples include tin compounds such as dibutyltin dilaurate,

tin octylate, dibutyltin dimaleate, dibutyltin diacetyl acetonate and reaction products of dibutyltin oxide and esters of phthalic acid, acetyl acetonate complexes such as aluminum-tris-acetyl acetonate, zirconium tetra-acetyl acetonate or titanium tetra-acetyl acetonate, amine compounds and their derivatives such as butylamine, dibutylamine, octylamine, monoethanolamine, diethanolamine, triethanolamine, morpholine, diethylenetriamine, triethylenetetramine, etc., as well as mixtures of the crosslinking catalysts mentioned above. The inventive sealing compound may also be prepared without using crosslinking catalysts, however.

Adhesion promoter substances may be selected from the group of silanes, e.g., 3-glycidyloxypropyl trialkoxysilane, 3-aminopropyl trialkoxysilane, *n*-aminoethyl-3-aminopropyl trialkoxysilane, 3-methacryloxypropyl trialkoxysilane, vinyl trialkoxysilane, isobutyl trialkoxysilane, 3-mercaptopropyl trialkoxysilane as well as those from the group of silicate esters, e.g., tetra-alkyl orthosilicates and from the group of metallates, e.g., tetra-alkyl titanates or tetra-alkyl zirconates as well as mixtures of the adhesion promoter substances listed above. The inventive sealing compound may also be prepared without the use of adhesion promoter substances, however.

The stabilizers may be antioxidants of the sterically hindered phenol type (e.g., tetrakis[methylene-3-(3,5-di-*tert*-butyl-4-hydroxyphenyl) propionate]methane or of the sulfur-based antioxidant type, such as mercaptans, sulfides, polysulfides, thiourea, mercaptals, thioaldehydes, thioketones, etc., and they may be used either alone or in mixtures.

Examples of reinforcing and non-reinforcing fillers include carbon black, precipitated or pyrogenic silica, precipitated or ground (or surface treated) chalk, clay, kaolin, talc, quartz, titanium dioxide, glass fibers, carbon fibers, graphite, aluminum powder and zinc powder and mixtures thereof.

Water binding fillers such as silica gel, calcium oxide or zeolites with defined pore diameters may be used as the desiccants. Zeolites with a pore diameter of 3 Å are especially preferred; they are capable of very effectively binding water but they are not suitable for uptake of gases such as nitrogen, oxygen or argon, and therefore they do not lead to increased deflection of the pane of glass when there are temperature fluctuations. Mixtures of the desiccants may also be used.

The sealing compound is produced by mixing the silane-modified butyl rubber with other ingredients described above in a suitable apparatus (compounder, twin-screw extruder or single-screw extruder) with high shearing. Optionally the mixing may also be performed under a protective gas or *in vacuo*. It is advantageous that the same unit in which the silane modification of the butyl rubber has been performed can also be used for this process step.

In another embodiment of this invention, a two-component sealing compound may also be used in which the silane-modified butyl rubber is used together with other ingredients to form a first component and the catalyst together with fillers, tackifying additives or other thermoplastic polymers forms the second component. The mixture of the two components is then prepared in a mixing apparatus shortly before application of the sealing compound to the pane of glass.

Because of the cooling after application, this sealing compound achieves the natural strength required for handling immediately after fabrication of the insulating glass unit, in contrast with two-component sealing compounds known in the past which are initially in the form of a paste and develop their natural strength only as a result of the crosslinking reaction. In this embodiment, the better stability of the two components in storage is another advantage.

In another embodiment, the second component may be water or fillers that release water (e.g., hydrogen carbonates [bicarbonates], hydrogen sulfates [acid sulfates], phosphates, phosphites, hydroxides, boric acid, clays, etc.) which accelerate the crosslinking of the sealing compound and thus advantageously accelerate the curing. The mixture of the two components is preferably performed here again just before application of the sealing compound to the pane of glass.

The inventive sealing compound may be used for production of multilayer insulating glass without any additional spacer and then performs the following functions:

- ! spacer between the glass sheets of the multilayer insulating glass;
- ! matrix for the desiccant for drying the volume formed by the sheets of glass and the edge structure in the insulating glass unit;
- ! barrier for water vapor which tends to penetrate through the sealing compound into the insulating glass unit from the outside;
- ! barrier for gases which tend to escape from the insulating glass unit through the sealing compound to the outside;
- ! elastic edge sealing.

The natural strength of the sealing compound which is required for handling the finished insulating glass unit is achieved immediately after fabrication by cooling after application. This is an advantage in comparison with the two-component sealing compounds based on polysulfide or polyurethane which are traditionally used and which cure at room temperature but react only in the course of a crosslinking reaction to be converted from a pasty compound to a crosslinked body having dimensional stability. The silane modification of the sealing compound according to this invention permits first a better adhesion to the polar glass surface and second a crosslinking initiated by moisture and thus a greater elasticity of the sealing compound.

The multilayer insulating glass produced with the sealing compound according to this invention may also be filled primarily with at least one noble gas (e.g., argon, krypton or xenon), at least one heavy gas (e.g., sulfur hexafluoride) or mixtures of the two types of gas in order to improve the thermal insulation and sound insulation properties.

The method for producing multilayer insulating glass using the inventive sealing compound is divided into the following steps:

- ! application of the inventive sealing compound to the edge area of a sheet of glass by means of a suitable apparatus (e.g., extruder or drum pump);

- application of a second pane of glass to the sealing compound with the same coverage;
- pressing the insulating glass unit to the predetermined thickness;
- optional filling of the volume in the insulating glass unit with noble gas and/or heavy gas;
- optionally repeating the process steps when producing multilayer insulating glass having more than two panes of glass;
- when using a two-component sealing compound, mixing the two components is provided as an additional first process step.

The multilayer insulating glass produced in this way may be used as construction insulating glass, as vehicle insulating glass or as marine insulating glass.

The sealing compound according to this invention for production of multilayer insulating glass without requiring an additional spacer has the advantage of improved resistance to weathering in comparison with the sealing compounds traditionally used in the production of insulating glass based on polysulfide or polyurethane (especially with respect to UV radiation and high atmospheric humidity) with a good water vapor barrier effect at the same time and good adhesion to glass. In addition, it allows inexpensive production of multilayer insulating glass while eliminating several production steps in comparison with traditional processes. Furthermore, damaged or useless insulating glass units produced by this process are more easily separated into the individual components, i.e., glass and sealing compound, than the units produced by traditional systems where the edge sealing is designed as a composite material; these individual components, i.e., glass and sealing compound, can in turn be sent for recycling.

The products containing the inventive sealing compound for production of multilayer insulating glass have the advantage that the crosslinking is induced by moisture, either penetrating from the outside due to diffusion into the sealing compound or already being present in the sealing compound. In the crosslinking of condensed Si-OH groups to form water, if X is a hydrolyzable group, the Si-OH groups are first formed by hydrolysis, whereby it is possible to rely on natural ambient moisture or additives which release water. This crosslinking may be performed before or after further use. This results in a desired increase in plasticity. It is also advantageous that the inventive sealing compound is characterized by a great adhesion to polar surfaces.

The inventive sealing compound and/or products containing the inventive sealing compound can be shaped by suitable methods for further use. This may be accomplished, e.g., by an extruder or by a pump with a downstream nozzle in the case of an adequately high temperature, e.g., of 100°C to 200°C (in this temperature range, the viscosity of the sealing compound is low enough). The body shaped in this way may be crosslinked to advantage under the influence of moisture.

Various embodiments of the claimed sealing compound are described in greater detail in the following examples. All the percentage amounts given are percent by weight (wt%) unless otherwise indicated.

Examples 1 through 3 and Comparative Examples A and B illustrate the reaction of butyl rubber with a mercapto-functional silane with and without free radical forming agents. A moisture-crosslinking polymer is then formed under the reaction conditions according to this invention (Examples 1 through 3).

Examples 1 through 3 (reactions of butyl rubber with mercapto-functional silane and free radical forming agent)

Mixtures of butyl rubber, 3-mercaptopropyl trimethoxysilane, azobisisobutyronitrile and stabilizer (this only in Examples 1 and 3) were fed into a heated compounder (see Table 1 for amounts used and temperatures).

Table 1

Reaction of butyl rubber with mercapto-functional silane (Examples 1 through 3)

	Example 1	Example 2	Example 3
Butyl rubber	50.6 g (85.2%)	100.0 g (92.3%)	150.1 g (91.9%)
3-Mercapto-propyltrimethoxysilane	7.9 g (13.3%)	7.7 g (7.1%)	11.5 g (7.0%)
Azobisisobutyronitrile	0.66 g (1.1%)	0.67 g (0.6%)	1.0 g (0.6%)
Stabilizer	0.25 g (0.4%)	-	0.75 g (0.5%)
Temperature (°C)	150	150	100

After two hours of compounding, the compounded mass was removed and a sample of 0.5 g of the mixture was dissolved in 10 mL hexane. After adding 0.25 mL of a 5% hexane solution of dibutyltin dilaurate, the solution was poured into water. With all the samples, the resulting film after evaporating the hexane was no longer soluble in hexane. This shows that the silane in the inventive Examples 1 through 3 was covalently bonded to the polymer and the polymer was moisture crosslinking.

Comparative Examples A and B (reaction of butyl rubber with mercapto-functional silane without a free radical forming agent)

Mixtures of butyl rubber, 3-mercapto-propyltrimethoxysilane and stabilizer were mixed in a heated compounder (see Table 2 for starting amounts and temperatures).

Table 2

Reaction of butyl rubber with mercapto-functional silane without free radical forming agent (Comparative Examples A and B)

	Comparative Example A	Comparative Example B
Butyl rubber	50.0 g (92.5%)	50.0 g (92.5%)
3-Mercapto-propyltrimethoxysilane	3.8 g (7.0%)	3.8 g (7.0%)
Stabilizer	0.25 g (0.5%)	0.25 g (0.5%)
Temperature (°C)	100	150

After mixing for two hours, the compounded mass was removed and the crosslinkability was tested with a 0.5 g sample as described in Example 1. The films thus obtained were soluble in hexane and petroleum spirit; in other words, there was no development of a covalent bond between silane and the polymer and the resulting polymer was not crosslinkable in moisture.

Comparative Examples C, D and E (reaction of butyl rubber with vinyl silane and free radical forming agent)

With these comparative examples, it will be shown that a moisture crosslinkable polymer could not be obtained by reacting butyl rubber with a vinyl silane under the influence of various free radical initiators.

Mixtures of 50.0 g (93.1%) butyl rubber, 0.25 g (0.5%) stabilizer, 0.45 g (0.8%) free radical forming agent
(Comparative Example C: dibenzoyl peroxide;
Comparative Example D: dicumyl peroxide;
Comparative Example E: azobisisobutyronitrile)
and 3.0 g (5.6%) of vinyl trimethoxysilane were needed for three hours in a compounder at 100°C to 150°C. After removing the mixtures, they were tested for their crosslinkability as described in Example 1. The films thus obtained were completely soluble in hexane and petroleum spirit, i.e., they were not moisture crosslinkable. This shows that butyl rubber cannot be modified according to this invention with vinyl silane as a silane without any mercapto group even under the influence of a variety of free radical initiators and the desired moisture crosslinkable polymer is not obtained.

Silane-modified butyl rubber was mixed with other ingredients in the following examples to form sealing compounds according to this invention. By extrusion of these sealing compounds onto glass, adhesive test bodies (structure: glass/sealing compound/glass) having a layer

thickness of the sealing compound of 4 mm and an adhesive area of 49 mm × 15 mm were produced. The adhesive test bodies were exposed to various environmental influences and were subjected to tensile testing at a pulling rate of 100 mm/min for various periods of time (two to four weeks). The tensile force required to destroy the test objects per unit of adhesive area was measured. This tested the influence of the silane modification of butyl rubber according to this invention on the aging resistance of the bonded glass produced with the help of the inventive sealing compounds (containing silane-modified butyl rubber).

Example 4 (Mixing the silane-modified butyl rubber with other ingredients to form a sealing compound)

In a compounder preheated to 100°C, 96.0 g (31.8%) silane-modified butyl rubber (prepared according to Example 3) was compounded with 48.0 g (15.9%) non-reinforcing carbon black, 8.0 g (2.7%) pyrogenic silica, 60.0 g (19.9%) desiccant, 48.0 g (15.9%) ethylene-propylene copolymer (softening point 100°C) and 1.47 g (0.49%) stabilizer. After 15 minutes, 30.0 g (10.0%) low molecular polyisobutylene and 10.0 g (3.3%) ethylene-propylene copolymer (softening point 120°C) were added and compounded for ten minutes. Then 0.03 g (0.01%) dibutyltin dilaurate was added and the mixture was needed for five minutes more. After compounding for two hours *in vacuo*, the compounded mass was removed from the compounder. The test bodies produced with this sealing compound exhibited good adhesion properties even after hot water storage and UV-hot water storage (see Table 3 for results).

Comparative Example F (mixing non-silane-modified butyl rubber with other ingredients)

In a compounder preheated to 100°C, 96.0 g (31.8%) butyl rubber was compounded with 48.0 g (15.9%) non-reinforcing carbon black, 8.0 g (2.7%) pyrogenic silica, 60.0 g (19.9%) desiccant, 48.0 g (15.9%), ethylene-propylene copolymer (softening point 110°C) and 1.50 g (0.50%) stabilizer. After 15 minutes, 30.0 g (10.0%) low-molecular polyisobutylene and 10.0 g (3.3%) ethylene-propylene copolymer (softening point 120°C) were added and compounded for 10 minutes. After compounding for two hours *in vacuo*, the compounded mass was removed from the compounder. The test bodies produced with this sealing compound which was not according to the present invention were tested for their adhesive properties (see Table 3 results).

Comparative Example G (mixing non-silane-modified butyl rubber with silane and other ingredients)

In a compounder preheated to 100°C, 89.0 g (29.5%) butyl rubber was compounded with 48.0 g (15.9%) non-reinforcing carbon black, 8.0 g (2.7%) pyrogenic silica, 60.0 g (19.9%) desiccant, 48.0 g (15.9%) ethylene-propylene copolymer (softening point 110°C) and 1.47 g (0.49%) stabilizer. After 15 minutes, 7.0 g (2.3%) 3-glycidyloxy-propyltrimethoxysilane, 30.0 g (10.0%) low molecular polyisobutylene and 10.0 g (3.3%) ethylene-propylene copolymer (softening point 120°C) were added and the mixture was compounded for 10 minutes. Then 0.03 g (0.01%) dibutyltin dilaurate was added and the mixture was compounded for 5 more minutes. After

compounding for two hours *in vacuo*, the compounded mass was removed from the compounder. The test bodies produced with this sealing compound which is not according to the present invention were tested for their adhesion properties (see Table 3 for results).

Table 3

Adhesion values for test bodies according to Example 4 and according to Comparative Examples F and G under various storage conditions (values in MPa)

Storage	Example 4	Comp. Example F	Comp. Example G
14 days at room temperature	1.1 ∀ 0.1	1.2 ∀ 0.1	1.1 ∀ 0.1
7 days at room temperature, 7 days at 70°C, water	1.1 ∀ 0.1	0.1 ∀ 0.1 (a)	0.4 ∀ 0.1 (a)
7 days at room temperature, 7 days at 70°C, dry	1.0 ∀ 0.1	0.9 ∀ 0.1	0.9 ∀ 0.1
7 days at room temperature, 21 days of UV-water storage	1.3 ∀ 0.1	- (b)	0.4 ∀ 0.1

(a) one test body disintegrated during storage
(b) all test bodies disintegrated during storage

It can be deduced from the results presented in Table 3 that only the adhesion test bodies produced using the inventive sealing compound yielded good adhesion values under all the selected storage conditions, whereas the adhesion test bodies produced with the sealing compound not according to this invention had little or not resistance to water and UV radiation.

Example 5 (Mixing the silane-modified butyl rubber with other ingredients)

In a compounder preheated to 150°C, 2400 g (35.5%) silane-modified butyl rubber (prepared according to Example 3) was compounded with 845 g (12.5%) non-reinforcing carbon black, 122 g (1.8%) pyrogenic silica, 1014 g (15.0%) desiccant, 1400 g (20.7%) ethylene-propylene copolymer (softening point 110°C) and 33.1 g (0.49%) stabilizer. After 15 minutes, 950 g (14.0%) low molecular polyisobutylene was added and compounded for 10 minutes. Then 0.68 g (0.01%) dibutyltin dilaurate added and the mixture was compounded for 5 minutes more. After compounding for two hours *in vacuo*, the compounded mass was removed from the compounder. The test items produced with this sealing compound had good adhesion properties even after storage in hot water and UV water storage (see Table 4 for results).

Example 6 (Mixing the silane-modified butyl rubber with other ingredients)

In a compounder preheated to 150°C, 2000 g (25.9%) silane-modified butyl rubber (produced

according to Example 3) was compounded with 650 g (8.4%) highly reinforcing carbon black, 190 g (2.5%) pyrogenic silica, 1600 g (20.7%) desiccant, 385 g (5.0%) talc, 1230 g (16.0%) ethylene-propylene copolymer (softening point 110°C) and 37.8 g (0.49%) stabilizer. After 15 minutes, 485 g (6.3%) low molecular polyisobutylene, 850 g (11.0%) hydrogenated hydrocarbon resin and 285 g (3.7%) ethylene-propylene copolymer (softening point 120°C) were added and the mixture was compounded for 10 minutes more. Then 0.77 g (0.01%) dibutyltin dilaurate was added and the mixture was compounded for 5 minutes more. After compounding for two hours *in vacuo*, the compounded mass was removed from the compounder. The test body produced with this sealing compound showed good adhesion properties even after hot water storage and UV-water storage (see Table 4 for results).

Example 7 (Mixing the silane-modified butyl rubber with other ingredients)

In a compounder preheated to 150°C, 2100 g (26.7%) partially crosslinked silane-modified butyl rubber (produced according to Example 3) was compounded with 750 g (9.5%) highly reinforcing carbon black, 210 g (2.7%) pyrogenic silica, 1230 g (15.7%) desiccant, 1050 g (13.4%) ethylene-propylene copolymer (softening point 110°C) and 38.5 g (0.49%) stabilizer. After 15 minutes, 86.5 g (1.1%) 3-glycidyloxypropyl-trialkoxysilane, 1180 g (15.0%) low-molecular polyisobutylene, 835 g (10.6%) hydrogenated hydrocarbon resin and 380 g (4.8%) ethylene-propylene copolymer (softening point 120°C) were added and compounded for ten minutes. Then 0.9 g (0.01%) dibutyltin dilaurate was added and the mixture was compounded for five minutes more. After compounding for two hours *in vacuo*, the compound was removed from the compounder. The test bodies produced with this sealing compound had good adhesion properties even after warm water storage and UV-water storage (see Table 4 for results).

Comparative Example H (Traditional polysulfide sealant)

For comparison purposes, the test results of adhesion of a known polysulfide sealant for sealing multilayer insulating glass are shown in Table 4.

Comparative Example J (Traditional polyurethane sealant)

For comparison purposes, the test results of adhesion of a known polyurethane sealant for sealing multilayer insulating glass are shown in Table 4.

Table 4

Adhesion values for test bodies according to Examples 5 through 7 and Comparative Examples H and J (values in MPa)

Storage	Example 5	Example 6	Example 7	Comp.	Comp.

				Example H	Example J
14 days at room temperature	1.2 ∀ 0.1	1.3 ∀ 0.1	1.3 ∀ 0.1	1.2 ∀ 0.1	1.1 ∀ 0.1
7 days room temperature, 7 days at 70°C, water	1.3 ∀ 0.1	1.2 ∀ 0.1	1.1 ∀ 0.1	0.9 ∀ 0.1	0.9 ∀ 0.1
7 days room temperature, 7 days at 70°C, dry	1.1 ∀ 0.1	1.2 ∀ 0.1	1.4 ∀ 0.1	1.2 ∀ 0.1	1.1 ∀ 0.1
7 days room temperature, 21 days UV- water storage	1.3 ∀ 0.1	1.3 ∀ 0.1	1.3 ∀ 0.1	0.8 ∀ 0.1	0.9 ∀ 0.1

Table 4 shows that the adhesive bodies produced with the inventive sealing compounds have at least equally good adhesion values, in some cases even better adhesion values than those of adhesion test bodies produced with the known sealants.

Example 8 (Production and testing of multilayer insulating glass)

Strands of the dimensions 16 mm × 8 mm were produced by extrusion of the sealing compounds from Examples 4 through 7 and then were placed on the edge area of a pane of glass (having the standard test body dimensions of 350 mm × 500 mm. Then a second pane of glass was placed flush over the first and the insulating glass unit was pressed with a distance between them (interspace between the panes of glass): 12 mm.

All these multilayer insulating glass units were tested according to DIN 1286, Part 1 and were also subjected to additional tests such as constant storage (at 52°C and 100° relative atmospheric humidity), outdoor storage, alternating pressure/suction stress in a vacuum chamber, pressure loading, shearing, etc. These tests showed mainly the improved resistance of the inventive elastic sealing with respect to weathering influences in comparison with the two-component sealing compounds that cure at room temperature and have previously in most cases, e.g., polysulfide or polyurethane.

Patent Claims

1. Sealing compound for producing multilayer insulating glass with which the sealing compound serves as a spacer, characterized in that the sealing compound comprises a silane-modified butyl rubber.

2. Sealing compound according to Claim 1, characterized in that it has the following composition (amounts given in wt%):

(a) silane-modified butyl rubber	5-70%
(b) thermoplastic polymers	5-40%
(c) rubbers	0-40%
(d) tackifying additives	5-50%
(e) crosslinking catalysts	0-2%
(f) adhesion promoter substances	0-5%
(g) stabilizers	0.1-4%
(h) reinforcing and non-reinforcing fillers	5-60%
(i) desiccants	1-40%

3. Sealing compound according to Claims 1 and 2, characterized in that it has the following preferred composition (amounts given in wt%):

(a) silane-modified butyl rubber	10-60%
(b) thermoplastic polymers	7-35%
(c) rubbers	2-35%
(d) tackifying additives	6-40%
(e) crosslinking catalysts	0-1%
(f) adhesion promoter substances	0-3%
(g) stabilizers	0.2-3%
(h) reinforcing and non-reinforcing fillers	7-50%
(i) desiccants	5-35%

4. Sealing compound according to Claims 1 through 3, characterized in that the silane-modified butyl rubber is produced by reacting butyl rubber with at least one silane containing mercapto groups and also containing hydroxyl groups or hydrolyzable groups and with at least one free radical forming agent.

5. Sealing compound according to Claims 1 through 4, characterized in that the silane-modified butyl rubber is produced by reacting butyl rubber with at least one free radical forming agent and at least one silane that contains mercapto groups as well as hydroxyl groups or hydrolyzable groups and has the following structure:

[see source for formula]

wherein
A denotes $(CH_2)_m$ or R^3-NH-CO-NH-R^4 or R^3-O-R^4 or R^3-S-R^4 and
m stands for a number between 1 and 10, preferably 1 to 6, especially preferably 3, and
R^1, R^2 stands for a substituted or unsubstituted alkyl group with 1 to 20 carbons or a substituted or unsubstituted aryl group with 6 to 20 carbons or a substituted or unsubstituted aralkyl group with 7 to 20 carbons and
R^1 and R^2 may be the same or different, and
X stands for hydroxyl group or a hydrolyzable group, and when there are several X groups in a molecule, they may be the same or different, and
a stands for 0, 1 or 2 and
b stands for 0, 1, 2 or 3 and
the sum of a and b is no greater than 4 and
n is an integer between 0 and 19 and
R^3, R^4 stand for $(CH_2)_m$ or $(CH_2)_m$-NH-$(CH_2)_m$ and
R^3 and R^4 may be the same or different.

6. Sealing compound according to Claim 5, characterized in that
A = $(CH_2)_3$, a = 0, b = 0, n = 0, X = OCH_3, or
A = $(CH_2)_3$, a = 0, b = 0, n = 0, X = OEt, or
A = $(CH_2)_3$, a = 0, b = 1, n = 0, X = OCH_3, $R^2 = CH_3$.

7. Sealing compound according to Claims 1 through 6, characterized in that the silane-modified butyl rubber is produced by reacting butyl rubber with at least one silane that contains mercapto groups and also hydroxyl groups or hydrolyzable groups and with at least one free radical forming agent from the group of azo compounds, peroxide compounds or redox initiators.

8. Sealing compound according to Claims 1 through 7, characterized in that the silane-modified butyl rubber is produced by reaction at temperatures close to or above the decomposition temperature of the free radical forming agent used.

9. Sealing compound according to Claims 1 through 8, characterized in that the silane-modified butyl rubber is produced in a compounder, a single-screw extruder or a twin-screw extruder.

10. Sealing compound according to Claims 1 through 9, characterized in that the silane-modified butyl rubber is produced by reacting partially crosslinked butyl rubber with at least one silane that contains mercapto groups and also hydroxyl groups or hydrolyzable groups and with at least one free radical forming agent.

11. Two-component sealing compound according to one of Claims 2 through 10, containing as the first component the ingredients (a) through (c) and (f) through (h) and as the second component the crosslinking catalyst (e).

12. Two-component sealing compound according to one of Claims 2 through 10, containing as the first component the ingredients (a) through (h) and as the second component a material that contains or releases water.

13. Use of the sealing compound according to Claims 1 through 12 for producing multilayer insulating glass for construction purposes, for automotive engineering and for marine engineering.

14. Use according to Claim 13 in which the sealing compound serves as a multifunctional sealant and at the same time as a spacer, a desiccant matrix, a water vapor barrier and gas diffusion barrier and as an elastic edge sealing.

15. Use according to Claims 13 through 14 in which the volume in the interspace between the panes of glass is filled primarily with at least one noble gas or one heavy gas or a mixture of the two.

16. Use according to Claims 13 through 15 in which the sealing compound is applied to the edge area of a pane of glass, then a second pane of glass is placed over it to cover it exactly and then the insulating glass unit is pressed to the predetermined thickness and finally the volume in the interspace between the panes of glass is filled with at least one noble gas or a heavy gas or a mixture of the two.

17. Use according to Claims 13 through 16 for producing multilayer insulating glass with two or more panes of glass.

18. Use according to Claims 13 through 17 in which the components having the composition according to Claim 11 or 12 are mixed shortly before the sealing compound is applied to the panes of glass.

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